Optical Investigations of $La_{7/8}Sr_{1/8}MnO_3$

J. H. Jung,* K. H. Kim, H. J. Lee, J. S. Ahn, N. J. Hur, and T. W. Noh†

Department of Physics, Seoul National University, Seoul 151-742, Korea

M. S. Kim and J.-G. Park

Department of Physics, Inha University, Inchon 402-751, Korea

(February 1, 2008)

Abstract

We investigated temperature dependent optical conductivity spectra of $La_{7/8}Sr_{1/8}MnO_3$. Its phonon bending mode starts to be splitted near a structural phase transition temperature. With further cooling, strength of a mid-infrared small polaron peak, which is located at 0.4 eV, becomes increased. These temperature dependent changes in the phonon spectra and the polaron absorption peaks were explained by a phase separation and a percolation-type transition.

PACS number; 71.27.+a, 71.38.+i, 75.30.Kz, 78.20.Ci

There have been lots of attentions paid on physical properties of doped manganites due to their scientific interests as well as potential applications. For $x > 0.17$, $La_{1-x}Sr_xMnO_3$ (LSMO) shows a metal-insulator (MI) transition and colossal magnetoresistance (CMR) near a ferromagnetic (FM) ordering temperature, T_C. On the other hand, LSMO samples with $0.10 \leq x \leq 0.17$ display a series of intriguing transitions. It experiences a structural change at T_S ($> T_C$). Just below T_C, its temperature-dependent resistivity, $\rho(T)$, shows a metal-like behavior. However, it finally enters into an insulating state at T_P ($< T_C$).[1]

Using neutron diffraction measurements, Kawano *et al.*[2] showed that the LSMO samples with $0.10 \leq x \leq 0.17$ have two kinds of orthorhombic phases: one is the O' phase with static Jahn-Teller (JT) distortion, while the other is a pseudocubic phase, called the "O^* phase". In particular, they claimed that the x=1/8 sample exhibits $O^* \rightarrow O' \rightarrow O^*$ structural transitions at T_S and T_P. From neutron scattering measurements, Yamada *et al.*[3] observed appearance of satellite reflection peaks below T_P. To explain this intriguing phenomenon, they used a polaron/charge ordering model. They showed that, below T_P, undistorted (001) planes where holes construct a 2×2 square lattice alternate with $(MnO_2)^-$ planes containing a static JT ordering of the oxygen displacements as in the $LaMnO_3$ (001) planes. They also argued that the locking into the commensurate structure should occur for LSMO samples within a finite concentration range around the nominal value of x=1/8.

Later, Zhou *et al.*[4] claimed that a dynamic segregation into the hole-rich and the hole-poor regions should occur at T_S, even though such a dynamic process would remain undetected by the diffraction experiments. Moreover, in the temperature range of $T_P < T < T_C$, they argued that the metallic behavior is not due to itinerant-electron conduction but that it comes from three-manganese clusters.

Recently, a couple of groups suggested existence of a phase separation (PS) in the lightly doped LSMO samples. Yunoki, Moreo, and Dagotto[5] performed computational studies on the 2-orbital Kondo model with JT phonons. With a moderate electron-phonon coupling, a PS was found to occur between (i) a metallic spin-FM and orbital-disordered phase (caused by the double-exchange interaction) and (ii) an insulating spin-FM and orbital-antiferro

(AF) ordered phase. Endoh *et al.*[6] observed an AF-type orbital ordering of e_g electron below T_P in $La_{0.88}Sr_{0.12}MnO_3$ by an anomalous x-ray scattering experiment. Using an effective Hamiltonian where spin and orbital degrees were treated on an equal footing, they showed the PS between the spin-FM phases. However, they claimed that the insulating phase was caused by superexchange interaction between neighboring e_g spins, so that the JT distortion was irrelevant in the insulating phase.[7]

In spite of these interesting physical phenomena, optical properties of $La_{7/8}Sr_{1/8}MnO_3$ have not been investigated in detail.[8] In this paper, we report details of our optical investigations, including T-dependent changes in phonon spectra and infrared (IR) absorption features. These changes will be discussed in terms of a phase separation and a percolation-type transition.

A polycrystalline $La_{7/8}Sr_{1/8}MnO_3$ sample was prepared by the conventional solid-state reaction method.[9] X-ray powder diffraction measurement showed that the sample did not contain secondary phases, and electron-probe microanalysis indicated that its chemical composition was stoichiometric within an error bar of 1 %. The T-dependent resistivity was measured by the four-probe method and magnetization, M, was determined using a SQUID magnetometer.

We measured reflectivity spectra $R(\omega)$ from 0.01 to 30 eV. To obtain T-dependent $R(\omega)$ in the frequency region from 0.01 to 2.5 eV, we used a liquid He-cooled cryostat. Since there were less than 2 % changes near 2.5 eV, we attached the low temperature $R(\omega)$ smoothly with room temperature data above the frequency. Optical conductivity spectra $\sigma(\omega)$ were obtained using the Kramers-Kronig (KK) transformation. Details of reflectivity measurements and the KK analysis for other manganite samples were described earlier.[10] Spectroscopic ellipsometry measurements were performed independently to obtain T-dependent $\sigma(\omega)$ in a frequency region between 1.5 and 5.0 eV. The optical conductivity data obtained from the spectroscopic ellipsometry agreed quite well with those from the reflectivity measurements, demonstrating validity of our KK analysis.

The inset of Fig. 1 shows $\rho(T)$ of $La_{7/8}Sr_{1/8}MnO_3$. Measured values of $\rho(T)$ were quite

close to the reported values.[11] Around T_S ($\sim$ 260 K), a weak thermal hysteresis behavior was observed. In the paramagnetic state above T_C ($\sim$ 190 K), $La_{7/8}Sr_{1/8}MnO_3$ shows an insulating behavior. Just below T_C, the $\rho(T)$ curve shows a metal-like behavior. However, below T_P ($\sim$ 160 K), it shows an upturn, indicating a reentrance into an insulating state. At the low temperature region, $\rho(T)$ can be fitted reasonably well with the variable range hopping model: $\rho(T)=\rho_0 \exp(\frac{C}{k_B T})^{1/4}$.

Figure 1 shows $R(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$ at various temperatures. The sharp peaks in the far-IR region are originated from optic phonon modes. Around 0.03 and 0.06 eV, additional small peaks can be seen clearly at low temperatures, indicating appearance of other phonon modes. Even for the metal-like region of $T_P < T < T_C$, $R(\omega)$ do not show any upturn behaviors at the low frequency region. [Reflectivity of a typical metal shows a upturn behavior in the dc limit (i.e., $\omega \rightarrow 0$).] Below T_C, $R(\omega)$ increase sharply around 0.3 eV and decrease slightly around 1.5 eV. In the temperature region of 200 K $\leq T \leq$ 300 K, $R(\omega)$ remain nearly the same. Above 2.5 eV, broad peaks due to the interband transition between electronic levels can be observed.

Figure 2 displays $\sigma(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$ in the far-IR region. There are three main optic phonon modes at 280 K: the phonons around 160, 350, and 570 cm^{-1} are known as external, bending, and stretching modes, respectively.[9] Just below T_S, the bending mode starts to be splitted slightly, and there are weak signatures for additional phonon modes around 220, 280, and 490 cm^{-1}. The external and the stretching modes are shifted to higher frequencies by about 3 cm^{-1} without any apparent splitting. With further cooling, the bending mode splitting and the additional phonon modes become more clear. The bending mode splitting and the additional phonon modes seem to be related to the polaron/charge ordering. With such an ordering, its lattice unit cell becomes enlarged, so there should be changes in the phonon spectra. Similar changes were observed below charge ordering temperature in $La_{1.67}Sr_{0.33}NiO_4$.[12]

There are three important points which should be addressed in these phonon spectra. First, the optical conductivity spectrum at 15 K ($\ll T_P$) is quite different from that at 280

K ($> T_S$), even though both of the crystallographic structures in these temperature regions are the pseudocubic O^*.[2] This demonstrates that the local symmetries in these two phases should be different. Contrary to the claim by Endoh *et al.*,[6] the local JT distortion might persist below T_P, which is consistent with recent pulsed neutron diffraction measurements.[13] Second, the bending mode splitting starts to occur at T_S (neither T_P nor T_C), suggesting that the phonon spectra changes should be closely related to the structural change. Third, there is little change in the stretching phonon mode position. In the $La_{0.7}Ca_{0.3}MnO_3$, which shows a MI transition and CMR near T_C, the stretching mode shows a significant frequency shift of about 20 cm^{-1}.[9] This suggests that the metal-like region of $La_{7/8}Sr_{1/8}MnO_3$ might have characteristics different from those of CMR manganites below T_C.

The T-dependent $\sigma(\omega)$ in the mid-IR region are shown in Fig. 3. The sharp peaks below 0.1 eV are due to optic phonons. Above T_C, $\sigma(\omega)$ show a broad peak around 1.5 eV. Below T_C, the spectral weight (SW) around 0.4 eV increases significantly without apparent changes in its peak position. In a small polaron picture, the position of the mid-IR absorption should be about 4 times larger than corresponding polaron activation energy.[14] If this mid-IR peak can be attributed to the small polaron absorption, the activation energy in $La_{7/8}Sr_{1/8}MnO_3$ could be estimated to be about 0.1 eV, consistent with recent transport measurements.[15]

Note that the mid-IR features for $La_{7/8}Sr_{1/8}MnO_3$ are different from those of other CMR manganites, including $La_{0.825}Sr_{0.175}MnO_3$,[16] $Nd_{0.7}Sr_{0.3}MnO_3$,[17,18] and $La_{0.7}Ca_{0.3}MnO_3$.[19] First, in the CMR manganites, the SW in the high energy region is transferred to a Drude term, which is typically small, and a broad mid-IR peak. And, the position of the mid-IR peak shows a very strong T-dependence, which might be related to a crossover from small to large polaron.[19] However, the mid-IR peak position in $La_{7/8}Sr_{1/8}MnO_3$ is nearly independent of T. Second, even for the metal-like region of $T_P < T < T_C$, $\sigma(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$ do not show any Drude peak. This unusual behavior is consistent with the idea by Zhou *et al.* that the sample remains in an unconventional FM state without itinerant carriers.[4] They argued that the metallic behavior should be originated from three-manganese clusters, in which the hole was alternately at one of the two Mn^{3+} ions and was made mobile by

dynamic JT coupling to the oxygen vibrations between manganese atoms.

The inset in Fig. 3 shows $\sigma(\omega)$ measured at 85 K and 250 K by the spectroscopic ellipsometry in a photon energy region of $1.5 \sim 4.0$ eV: as T decreases, $\sigma(\omega)$ decrease slightly with a broad background. [A similar SW change in the visible-UV region was observed for a $La_{0.6}Sr_{0.4}MnO_3$ thin film using a transmission measurement.[20]] Within our experimental errors, the total change of the SW measured by the spectroscopic ellipsometry is close to that observed below 1.5 eV, and with an opposite sign. This fact suggests that a significant portion of the SW in the mid-IR region was transferred from those between 1.5 and 4.0 eV.

To investigate the SW changes quantitatively, we regarded $\sigma(\omega)$ as a sum of two components: $\sigma(\omega) = \sigma_{ms}(\omega)+\sigma_L(\omega)$, where $\sigma_{ms}(\omega)$ and $\sigma_L(\omega)$ represent contributions of midgap states and Lorentz oscillators, respectively. $\sigma_L(\omega)$ mainly come from interband transition of O $2p \rightarrow e_g$ and transitions from lower to upper Hund's rule split bands, i.e. $e_g^{\uparrow}(t_{2g}^{\downarrow}) \rightarrow e_g^{\uparrow}(t_{2g}^{\downarrow})$ and $e_g^{\downarrow}(t_{2g}^{\downarrow}) \rightarrow e_g^{\downarrow}(t_{2g}^{\uparrow})$.[20,21] [This notation indicates that the transitions occur between two e_g bands with the same spin but under different t_{2g} spin backgrounds.] Following our earlier works,[21] we initially fitted $\sigma(\omega)$ above 2.0 eV with a series of the Lorentz oscillator functions and then obtained $\sigma_{ms}(\omega)$ by subtracting $\sigma_L(\omega)$ from measured $\sigma(\omega)$.

The T-dependent $\sigma_{ms}(\omega)$ data are shown in Fig. 4, and they were analyzed with double peak structures. The strong peak near 1.5 eV, which will be called "Peak II", is nearly temperature independent. The peak near 0.4 eV, which will be called "Peak I", is quite weak at 210 K, but it becomes evident around 180 K (i.e. just below T_C). As T becomes lower, it becomes stronger. In our earlier doping dependent studies on optical properties of $La_{1-x}Ca_xMnO_3$,[21] similar double peak structures were observed. And, recent calculations by Yunoki, Moreo, and Dagotto[5] also predicted such double peak structures in $\sigma(\omega)$. Based on these works, we assigned Peaks I to a small polaron absorption.[22] With this assignment, the temperature dependence of Peak I can be explained qualitatively. Below T_C, t_{2g} spins are aligned to make it easier for an e_g electron to hop from a JT splitted Mn^{3+} site to an unsplitted Mn^{4+} site.

A fitting procedure with two Gaussian functions was used to find more quantitative

behaviors of this double peak structure. Strengths of the peaks, S_{ms}^{k} (k = I and II), were obtained by numerically integrating the corresponding Gaussian functions. Values of $S_{ms}^{\rm I}$ are shown as the solid squares in Fig. 5. [Values of $S_{ms}^{\rm II}$ are nearly T-independent within our error bars.] Above T_C, $S_{ms}^{\rm I}$ is small and nearly T-independent. Although the phonon spectra in Fig. 2 start to change at T_S, there is no apparent change in $S_{ms}^{\rm I}$ near this temperature. With cooling, $S_{ms}^{\rm I}$ sharply increases below T_C and becomes nearly saturated around 100 K. Although the T-dependence of $S_{ms}^{\rm I}$ is similar to that of M/M_S ($\equiv M^*$), where M_S is the saturated magnetization, there are a couple of differences which should be noted. First, above T_C, M vanishes, but $S_{ms}^{\rm I}$ seems not. Second, M saturates near T_P, but $S_{ms}^{\rm I}$ saturates around 100 K.

Without polaron/charge ordering, $S_{ms}^{\rm I}$ should depend simply on a number of available polaron absorption transitions. Suppose that an e_g electron at a JT splitted Mn^{3+} ion has an up-spin (which is parallel to M) and is located at a nearest neighbor site of a unsplitted Mn^{4+} ion. Since the polaron hopping process should conserve the spin, its strength should be proportional to $[1+M^*]^2/4$. For a down-spin, it should be proportional to $[1-M^*]^2/4$. Therefore, $S_{ms}^{\rm I}$ should be proportional to $[1+M^{*2}]/2$, which is the same as the behavior of the Drude peak strength predicted for the CMR manganites in the spin-split band picture.[23] However, the experimental values of $[1+M^{*2}]/2$, shown as the solid line in Fig. 5, do not agree with $S_{ms}^{\rm I}$. From our data, it is quite plausible to say that the photon-assisted polaron transfer matrix might be enhanced significantly at the polaron/charge ordered state, where M is already nearly saturated.

One of possible scenarios to explain the T-dependence of phonon spectra and $S_{ms}^{\rm I}$ is a percolation-type transition.[24] As suggested by Zhou *et al.*,[4] a dynamic phase segregation into the hole-rich and the hole-poor regions (i.e. formation of the polaron/charge ordered domains) might start to occur around T_S. Changes in the phonon mode will appear, and $S_{ms}^{\rm I}$ remains finite but small. At the temperature region of $T_P < T < T_C$, the FM ordering starts to occur and $S_{ms}^{\rm I}$ increase very rapidly. In this temperature region, a PS might occur between two spin-FM phases, i.e. a metallic orbital-disordered phase and an insulating

orbital-ordered phase, as suggested by Yunoki *et al.*[5] and by Endoh *et al.*[6] Near T_C, the metallic phase is favored because entropy promotes the orbital disordering and the carrier mobilities. However, below T_P, the insulating FM orbital-ordered phase becomes dominant, resulting the polaron/charge ordering observed by Yamada *et al.*[3] $S^{\rm I}_{ms}$ will increase until all of the sample becomes insulating in the polaron/charge ordered state. Even though this picture based on the PS and a percolation-type transition can explain most T-dependent changes in $\sigma(\omega)$ and $\rho(T)$, more studies are required to get a more clear picture.

In summary, we investigated optical conductivity spectra of $La_{7/8}Sr_{1/8}MnO_3$. The phonon bending mode starts to be splitted near a structural transition temperature. At lower temperature, the strength of the mid-infrared small polaron peak becomes increased. Even in the metal-like region, the Drude peak was not observed. These features supports that a phase separation and a percolation-type transition should occur in $La_{7/8}Sr_{1/8}MnO_3$.

We acknowledge valuable discussions with Prof. E. J. Choi, Prof. Jaejun Yu, and K. H. Ahn. This work are financial supported by the Ministry of Education through the Inter-University Center for Natural Science Research Facilities (BSRI-97-7401), by the Korea Science and Engineering Foundation through grant No. 971–0207-024-2 and through RC-DAMP of Pusan National University. Experiments at PLS were supported in part by MOST and POSCO.

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* Electronic mail address: hoon@phya.snu.ac.kr

† Also at LG Corporate Institute of Technology (LG-CIT), Seoul 137-742, Korea.

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[22] There have been different interpretations on the origin of the Peak II, which is located

around 1.5 eV: an interband transition from O $2p$ to Mn $3d$ levels (Ref. 16), an interatomic transition from a JT split Mn^{3+} lower level to a Mn^{3+} upper level (Ref. 18), and an intraatomic transition between the JT split Mn^{3+} levels (Ref. 21). Each assignment seems to have some strong and weak points. For more details, see Ref. 8. [However, the correct assignment of the origin of Peak II is not important in this paper.]

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FIGURES

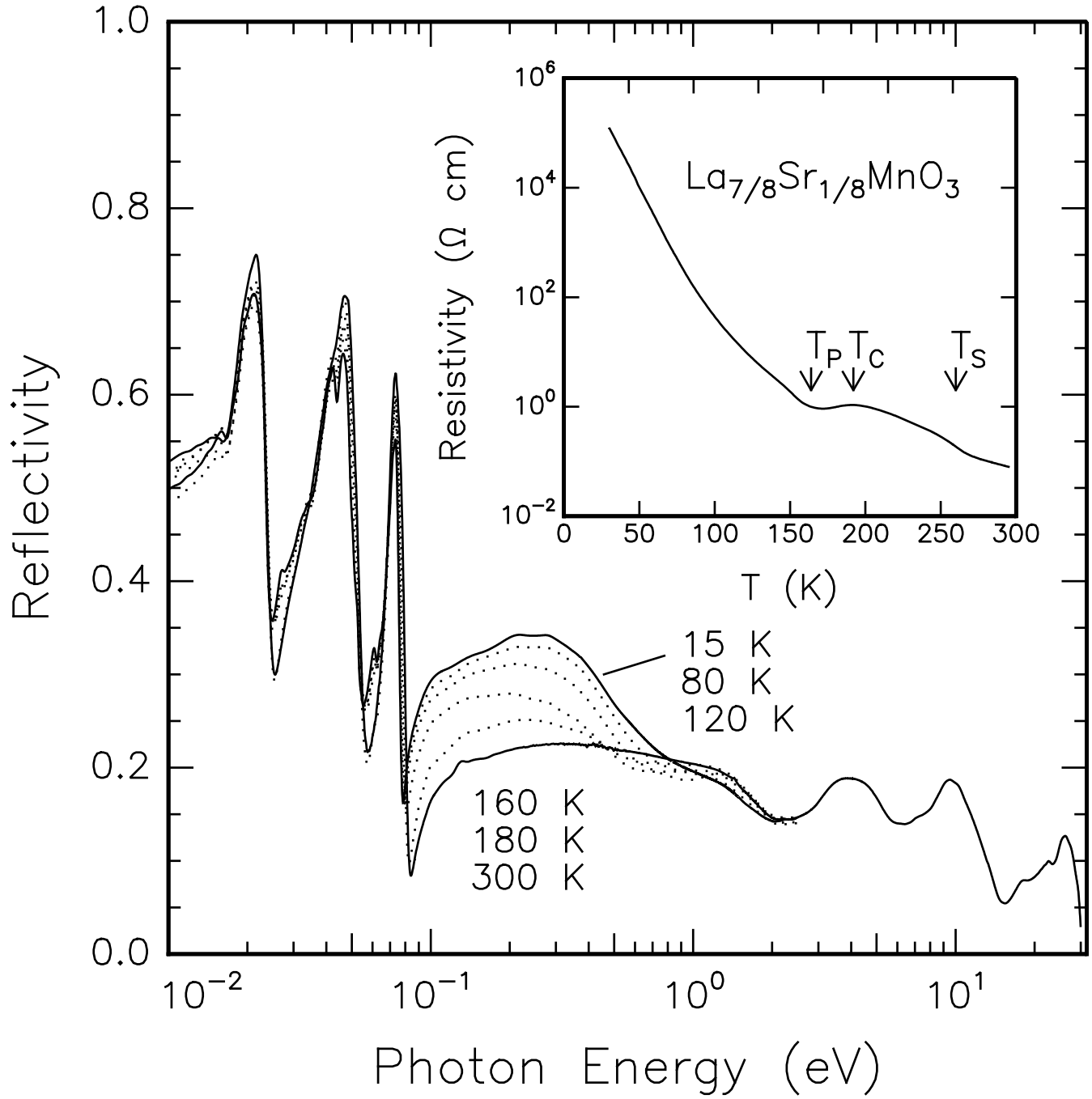
FIG. 1. Temperature dependent $R(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$. In the inset, $\rho(T)$ for the same sample is shown.

FIG. 2. Optic phonon modes of $La_{7/8}Sr_{1/8}MnO_3$. Positions of the external and the stretching phonon frequencies at 280 K are drawn as dotted lines.

FIG. 3. Temperature dependent $\sigma(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$ below 2.0 eV. In the inset, $\sigma(\omega)$ obtained from the spectroscopic ellipsometry are shown.

FIG. 4. The midgap state conductivity $\sigma_{ms}(\omega)$ of $La_{7/8}Sr_{1/8}MnO_3$. The solid circles and the dotted lines represent the experimental data and the Gaussian fitting lines, respectively.

FIG. 5. Values of $S^{\rm I}_{ms}$ in $La_{7/8}Sr_{1/8}MnO_3$. Predicted values of M^*, M^{*2}, and $[1+M^{*2}]/2$ were denoted as the dotted, the dot-dashed, and the solid lines, respectively.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15 K
80 K
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10^{2}
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Resistivity (Ω cm)
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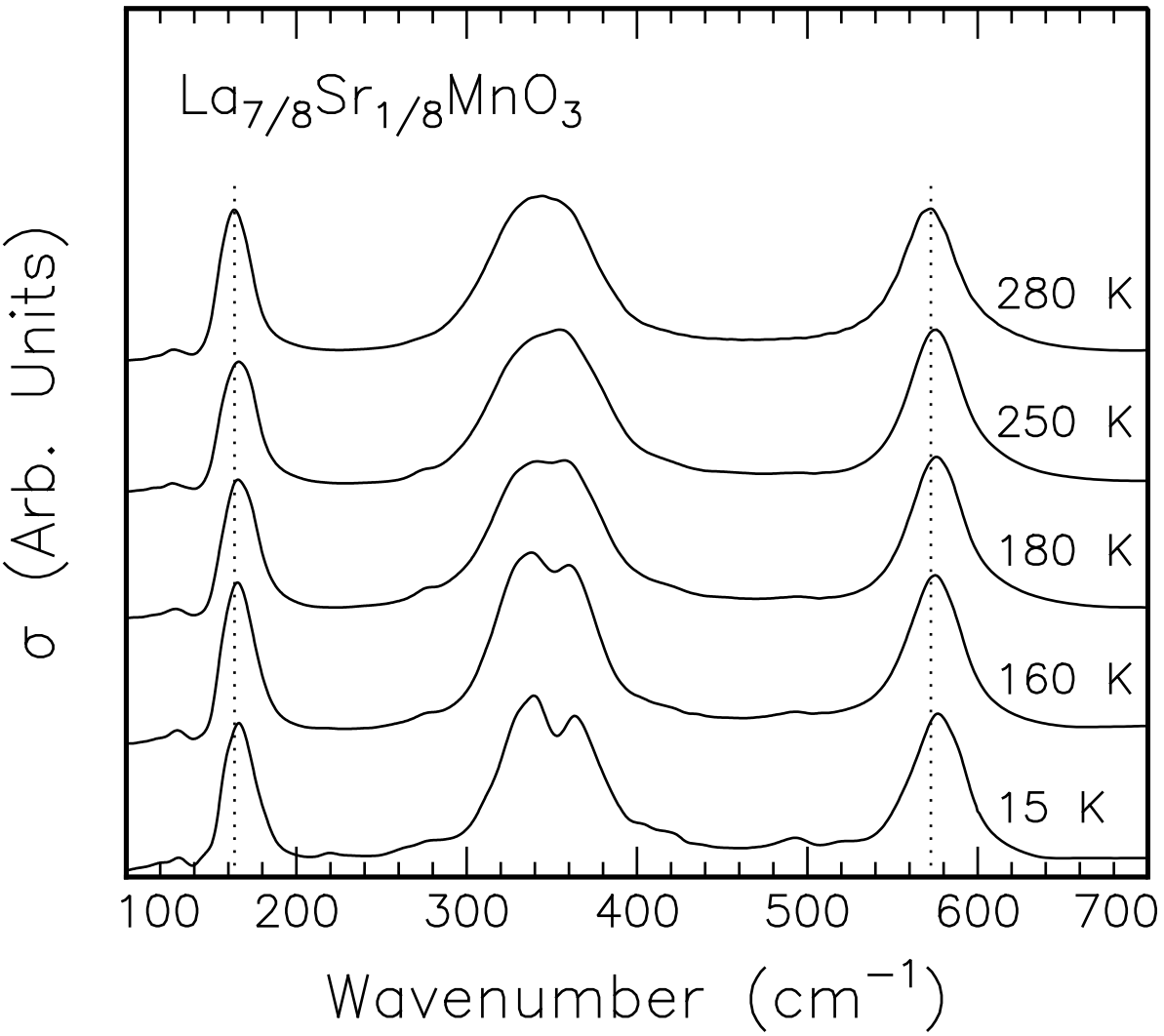
T (K)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15 K
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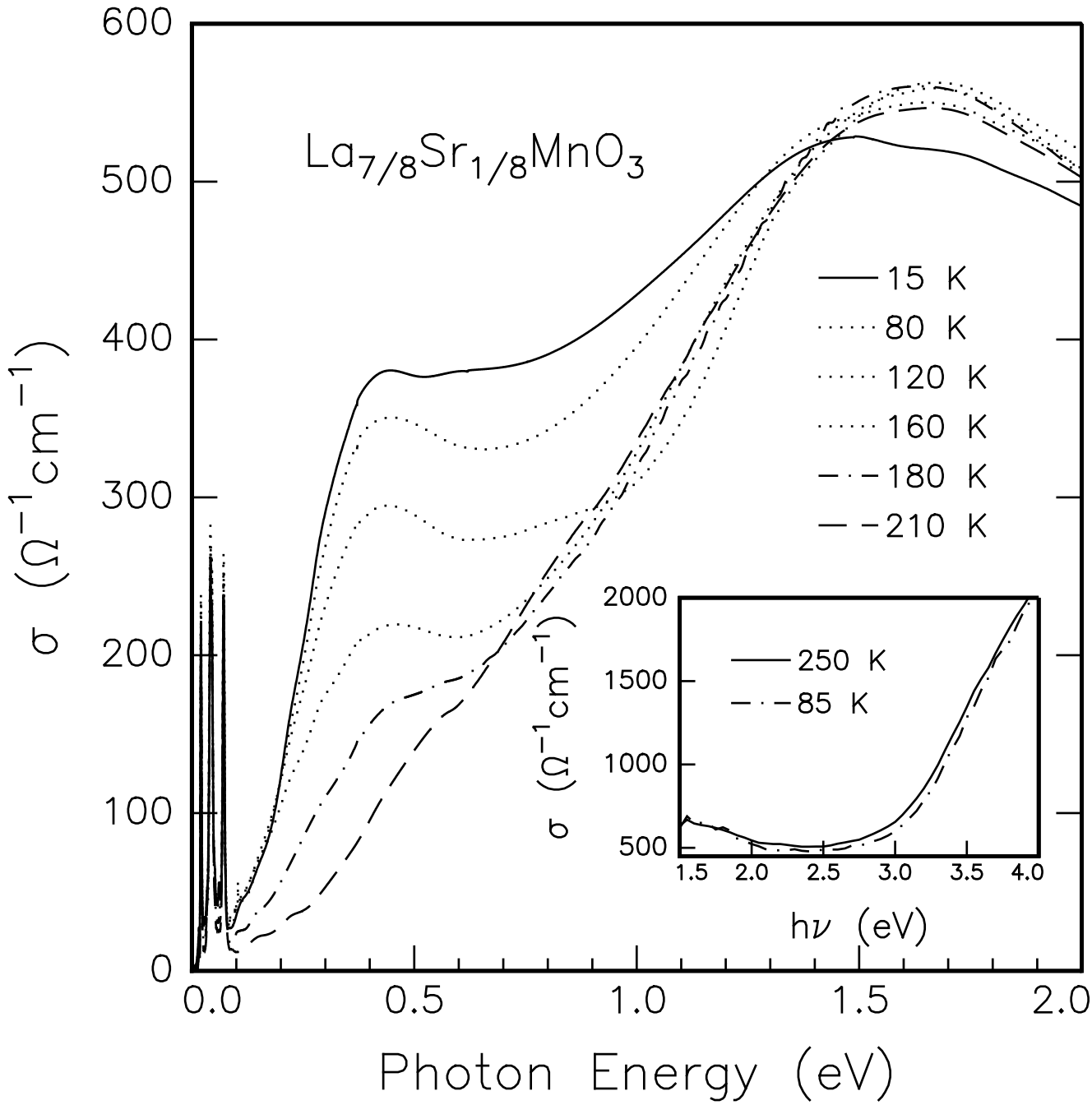
Wavenumber (cm−1)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σ (Ω−1cm−1)
Photon Energy (eV)
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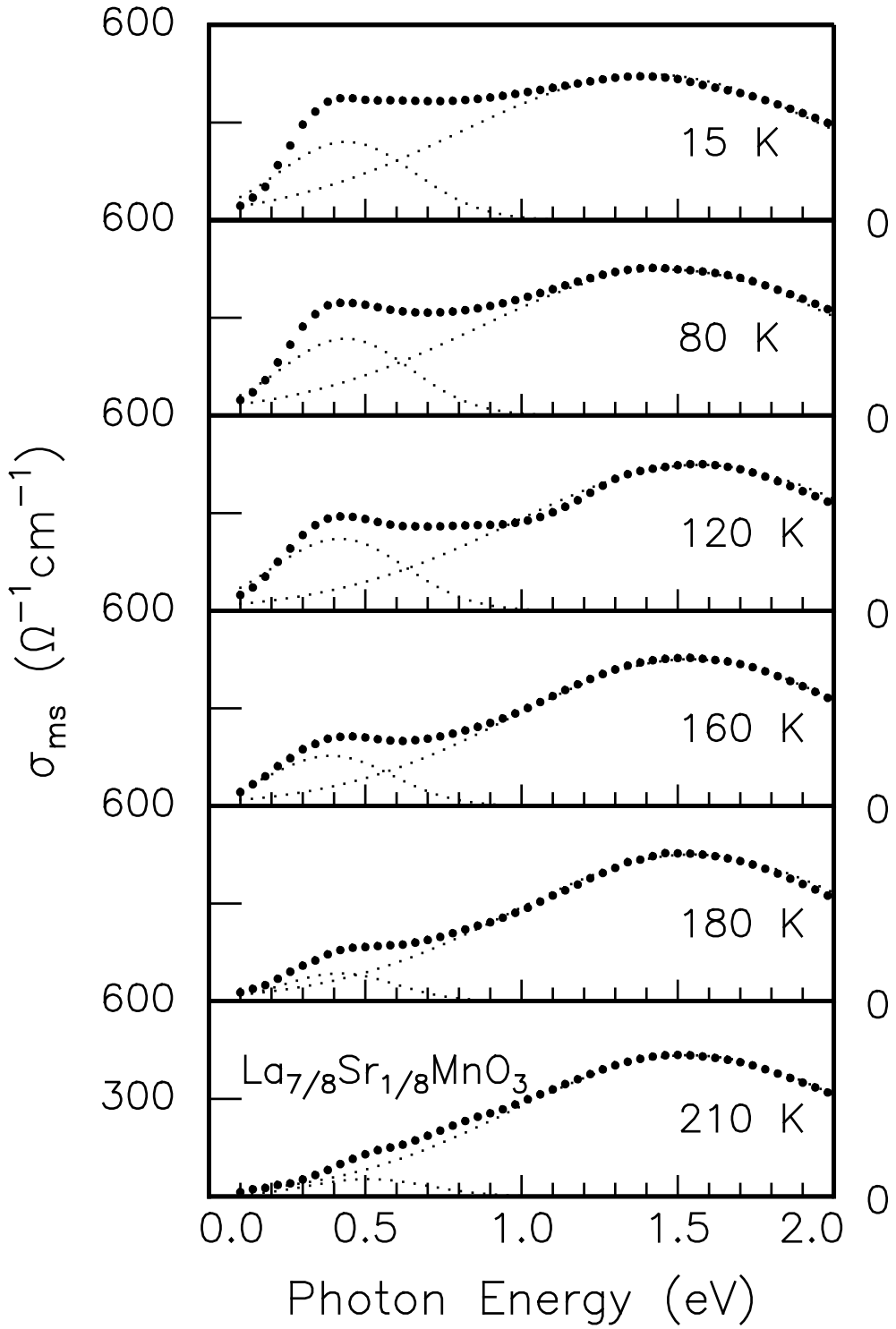
hν (eV)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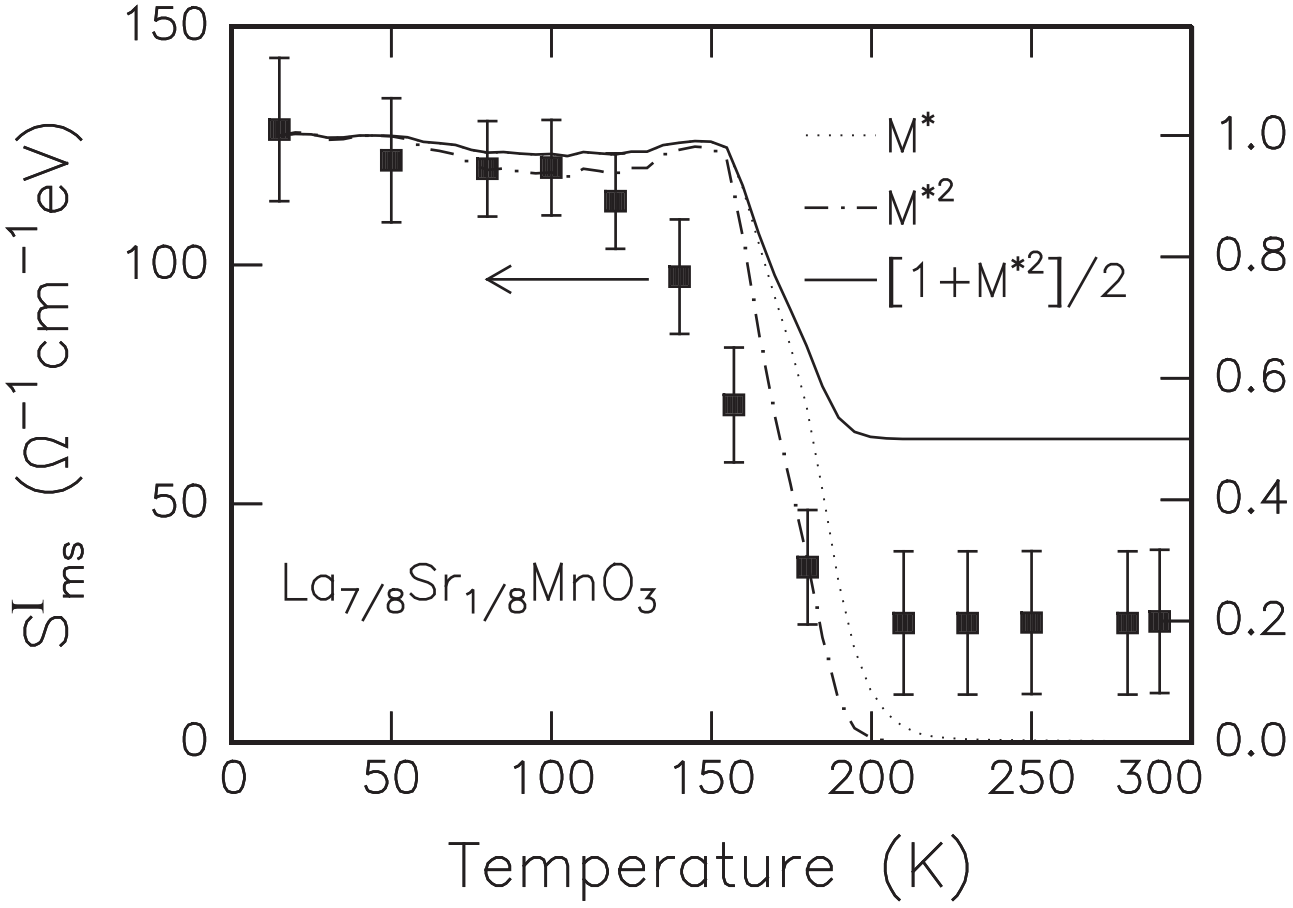
Photon Energy (eV)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S^I_{ms} ($\Omega^{-1}cm^{-1}eV$)
Temperature (K)
M*
M*2
[1+M*2]/2
La7/8Sr1/8MnO3